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Residues in Milk of Cows Fed Rations Containing Low Concentrations of Five Chlorinated Hydrocarbon Pesticides

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A feeding study involving 16 lactating dairy cows was carried out in which mixtures of 5 pesticides (heptachlor epoxide, dieldrin, endrin, lindane, and DDT) were fed at levels of approximately 0.05, 0.15, and 0.30 ppm of each pesticide based on total feed consumption. Analyses showed that heptachlor epoxide and dieldrin transferred to the milk in much higher concentrations than did the other pesticides. Next in order of concentration were endrin and lindane. Small but definite increases of DDT and TDE in the milk were noted with increased feeding levels of DDT, but the DDE concentration was apparently not affected by the feeding of DDT at these levels.

A number of studies have been made of the transfer of chlorinated pesticide residues from the feed to the milk of cows. Gannon, *et al.* (1) fed dieldrin to cows at levels of 0.1 to 2.25 ppm, and, using a colorimetric analytical procedure, found dieldrin in their milk. Gannon, *et al.* (2) also analysed milk from cows fed aldrin, dieldrin, heptachlor, DDT, and methoxychlor at levels of 1 to 7000 ppm and again found residues of the pesticides or their metabolites in the milk.

Zweig, *et al.* (3) fed DDT to cows at levels of 0.5, 1.0, 2.0, 3.0, and 5.0 ppm. Using a colorimetric method sensitive to about 0.01 ppm they did not find any DDT in milk from the cows fed at the 0.5 ppm level, but did find DDT in the milk of the cows fed at the higher concentrations.

Hardee, *et al.* (4) added heptachlor epoxide and Telodrin® to the feed of cows at levels of 5 and 20 ppb (0.005 and 0.020 ppm) and, using electron affinity gas chromatography, found measurable residues of each in the milk.

The present study was undertaken to provide additional information on the transfer of chlorinated pesticide residues from feed to milk, when fed at low concentrations. The pesticides used were lindane, heptachlor epoxide, dieldrin, *p,p'*-DDT, and endrin. These five were chosen because of their frequent occurrence in dairy animal feeds and milk and/or because of their toxicity. Feeding levels were set at 0.05, 0.15, and 0.30 ppm on the basis of anticipated total feed consumption.

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Procedure

Sixteen Holstein cows, all first lactation animals and in the 100th–200th day of lactation, were divided into 4 similar groups of 4 cows each. All animals were fed a diet consisting of approximately 20½% grain concentrate, 16% hay, 8½% hay pellets, and 55% corn silage. The daily feed consumption of each cow was determined during a 2-week period prior to the start of the study, and this figure was used in calculating the amount of pesticide to be added. The 4 cows in Group A were used as controls, while during the feeding period Group B cows were fed an added 0.05 ppm, Group C, 0.15 ppm, and Group D, 0.30 ppm of each of the 5 pesticides.

An alcohol solution of the five pesticides was added to the grain ration of each cow at the morning and evening feeding. After the entire grain ration was consumed, the cow was given the rest of its feed and any remaining unconsumed feed was removed and weighed. The entire grain ration was always consumed but some of the roughage was at times rejected. Since the feed consumption varied somewhat during the course of this study, the actual level of added pesticides in ppm was not identical for all cows in each group. Table 1 shows the body weight, average daily feed consumption, actual level of added pesticides, and average daily milk production with fat content for each cow.

A sample of each feed ingredient was taken at each feeding, and weekly composites of each ingredient were analyzed for pesticide residues. Table 2 shows the pesticide content of the total feed calculated from the analyses of the individual components.

Milk samples from each cow were collected

Table 1. Data for individual cows

	Cow No.	Body Weight, lb	Average Feed Consumption, lb/day	Level of Added Pesticides, ppm	Average Milk Production, lb/day	Milk, Average % Butterfat
Group A: Controls	670	1148	66	0	27.3	4.6
	600	1104	58	0	28.4	4.7
	607	1130	70	0	36.0	4.2
	668	1020	61	0	35.0	4.4
	Group Av.	1102	64	0	31.7	4.5
Group B: 0.05 ppm Feeding Level	677	1148	60	0.057	41.4	4.2
	660	1070	46	0.051	28.5	4.1
	459	1080	54	0.051	31.1	3.7
	662	1004	64	0.051	38.5	4.2
	Group Av.	1100	56	0.052	34.0	4.0
Group C: 0.15 ppm Feeding Level	664	1250	63	0.139	33.3	4.1
	402	1082	64	0.142	33.2	4.5
	673	1018	65	0.139	37.6	4.1
	665	1138	61	0.146	30.6	4.0
	Group Av.	1123	64	0.142	35.0	4.2
Group D: 0.30 ppm Feeding Level	654	1056	66	0.306	35.8	3.6
	457	1140	64	0.288	47.0	4.0
	606	1140	70	0.249	28.7	4.4
	659	1000	60	0.318	19.8	4.3
	Group Av.	1100	66	0.302	32.8	4.1

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Table 2. Pesticide content in ppm of total feed before addition of pesticides (calculated from analyses of individual feed components—weekly composites)

Pesticide	Prefeeding		Feeding Period					Post-Feeding		
	1st Week	2nd Week	1st Week	2nd Week	3rd Week	4th Week	5th Week	1st Week	2nd Week	3rd Week
Heptachlor epoxide	0.005	0.006	0.006	0.004	·	0.001	—	0.002	—	Not Analyzed
Dieldrin	0.003	0.002	0.003	0.005	0.003	0.003	0.001	0.003	0.001	
Endrin	—	—	—	0.002	—	0.002	—	—	<0.001	
Lindane	—	<0.001	<0.001	0.001	<0.001	0.002	0.001	—	<0.001	
o,p'-DDT	0.001	0.002	0.004	0.004	0.010	0.014	0.022	0.020	0.009	
p,p'-DDT	0.007	0.009	0.010	0.013	0.021	0.015	0.023	0.010	0.011	
p,p'-TDE	—	—	—	0.002	0.002	0.003	0.002	0.002	0.002	
p,p'-DDE	0.003	0.005	0.004	0.004	0.004	0.006	0.004	0.004	0.004	

every Sunday and Wednesday. Milk from the morning and evening milkings was composited, fat content determined, and the milk then analyzed for pesticide residues. Milk samples were extracted and cleaned up by the rapid procedure of Onley (5), feed samples by the procedure of Mills, *et al.* (6). All samples were examined by the electron capture gas chromatographic procedure of Burke and Giuffrida (7), which is capable of detecting as little as 0.001 ppm of these pesticide residues. Most of the milk samples were also checked by the microcoulometric gas chromatographic procedure of Burke and Johnson (8) and the thin-layer chromatographic method of Kovacs (9). These latter determinations were made on the sample extract used for the electron capture gas chromatographic determinations. Analyses were made for the entire duration of the study, which consisted of a 2-week prefeeding period, a 5-week feeding period, and a 3-week post-feeding period. Thus, a total of 320 milk samples were analyzed, each being examined for 7 pesticide residues (the 5 pesticides that were fed, as well as TDE and DDE).

Results and Discussion

Although all analyses were made on milk from individual cows, the results for each sample day have been averaged by groups of four according to feeding level. Tabulations have been based on the electron capture gas chromatographic determination except for about 1% of the results; in these cases because of interference or obvious errors, the electron capture result was discarded and the microcoulometric gas chromatographic and/or thin-layer chromatographic result was used. No corrections have been made in any of the reported results for the residues found in the milk of the control animals.

During the prefeeding period, residues found in the milk of all groups, including the controls, were about the same.

Figures 1–4 show changes with time for the concentration in the milk of residues of heptachlor epoxide, dieldrin, endrin, and lindane, plotted on semilogarithmic graphs. It is apparent that for any one feeding level, considerably more heptachlor epoxide and dieldrin carried through to the milk than did the other pesticides. At the 0.30 ppm feeding level, heptachlor epoxide in the milk had reached a concentration of about 0.14 ppm and appeared to be still increasing at the end of 35 days' feeding.

Residues of both heptachlor epoxide and dieldrin, averaging about 0.005 ppm each, were found in all samples of milk from the control animals. There appeared to be a gradual decrease from about 0.006 ppm during the prefeed period to about 0.004 ppm at the end of the feeding period. It is possible that these residues were not specifically related to the intake during this study, but

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could have been due, at least in part, to earlier ingestion of feeds that may have been contaminated.

Residues of endrin and lindane were found in the milk at all three feeding levels, although at lower concentrations than heptachlor epoxide and dieldrin. No endrin or lindane was found in the milk of the control animals.

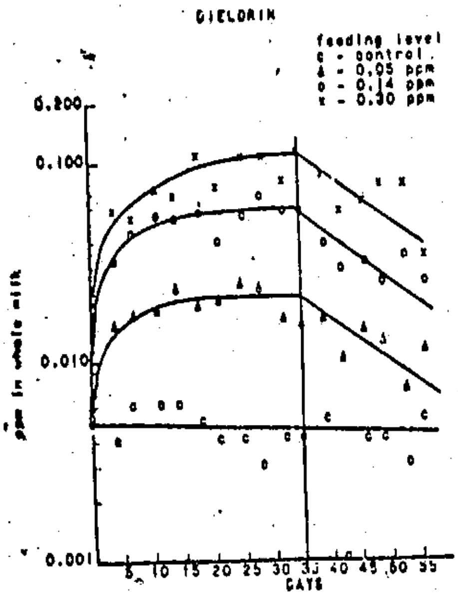

Fig. 1—Changes in concentration of heptachlor epoxide in milk with time.

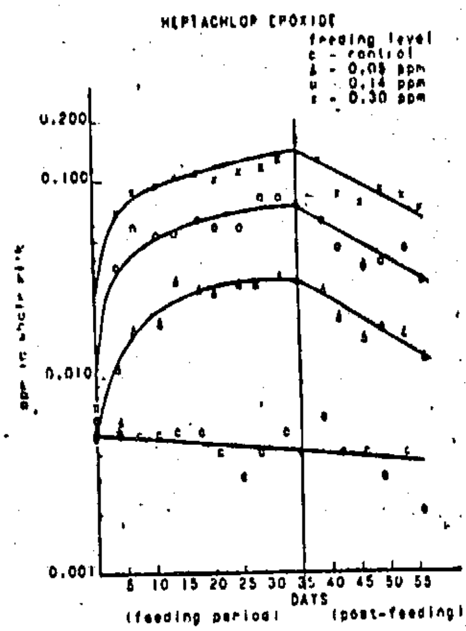

Fig. 2—Changes in concentration of dieldrin in milk with time.

Table 3 lists the "plateau" concentration for each of the residues in milk. Except for heptachlor epoxide at the two higher feeding levels, the concentration of each residue in milk had apparently reached a maximum by the end of the 35-day feeding period.

The residues of DDT, DDE, and TDE in the milk were so low that graphs would not be informative. As shown in Table 3, there were small but definite increases in residues of both *p,p'*-DDT and TDE at the higher feeding levels. However, the highest residues found were still under 0.01 ppm.

DDE residues averaged about 0.004 ppm for the controls and for all feeding levels throughout the entire study. Apparently the pesticides added to the feed did not affect the DDE concentration in the milk.

Table 3. "Plateau" levels: pesticide residues in milk at end of feeding period, ppm

Pesticide	Feeding Levels, ppm 0	0.052	0.142	0.302
Heptachlor epoxide	0.004	0.031	0.072*	0.14*
Dieldrin	0.004	0.021	0.058	0.11
Endrin	0	0.004	0.010	0.018
Lindane	0	0.002	0.006	0.015
p,p'-DDT	0.002	0.004	0.004	0.007
p,p'-TDE	0.001	0.002	0.003	0.004
p,p'-DDE	0.004	0.004	0.004	0.004

* Appeared to be still increasing at end of feeding period.

Feed consumption and milk production were very uniform for the entire duration of this study. Although the individual cows showed some daily variation, no changes could be correlated with the level of pesticide intake.

It should be pointed out that the results presented were obtained by feeding mix-

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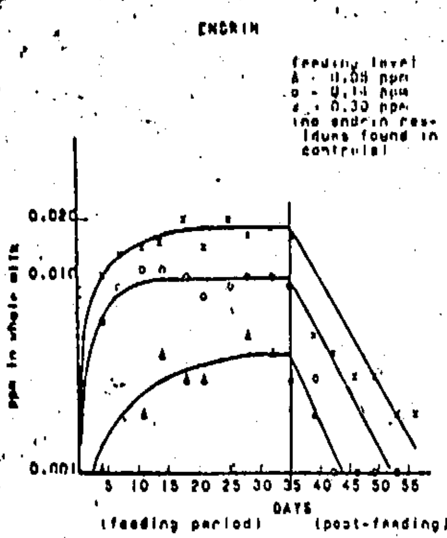

Fig. 3—Changes in concentration of endrin in milk with time.

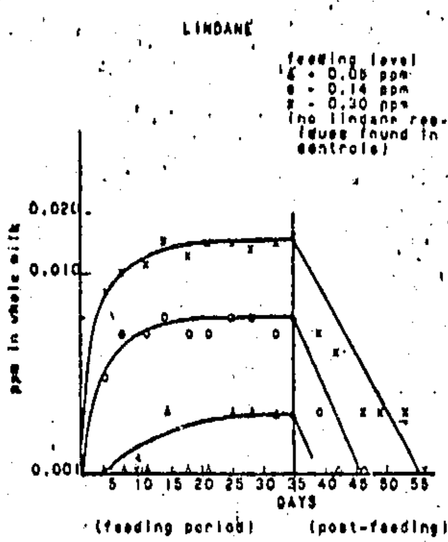

Fig. 4—Changes in concentration of lindane in milk with time.

tures of five pesticides. It is possible that the transfer rate of each individual pesticide was influenced by the presence of the other four, and that different results might be obtained by feeding only one pesticide to a cow. Also, although the curves in Figs. 1–4 were drawn to the end of the study period, the 3-week post-feeding period during which milk samples were analyzed is too short to permit accurate determination of the rates of decay.

Summary

This study has shown that even very low concentrations of heptachlor epoxide or dieldrin in a cow's feed will result in measurable residues in its milk. To a lesser degree, this is also true of endrin and lindane, and to a still smaller degree of DDT and its metabolite, TDE.

Acknowledgments

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